Hygrothermal Characterisation and Diffusion Studies on Glass Epoxy Composites

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ABSTRACT: Hygrothermal studies were carried out on a bidirectional (BID) glass fabric epoxy (LY 5052) composite specimens of $V_f = 0.5$ for three different cases of conditioning viz. 45°C/85% RH, 45°C/95% RH and 45°C/immersion (100% RH) until saturation was reached. Specimens of different test configurations [(Longitudinal Tensile (LT), Transverse Tensile (TT), Longitudinal Compression (LC), Transverse Compression (LC) and Inplane Shear (IPS)] were conditioned and their diffusion parameters estimated. Fickian diffusion behavior was, in general, observed for all these specimens.

INTRODUCTION

FIBER REINFORCED COMPOSITES provide immense application potential in the building of aircraft, chemical plant equipment and other industrial uses. In most of the applications these are exposed to varied temperature and moisture combinations. It is important to experimentally determine the extent of moisture absorption in polymeric composites and hence corresponding diffusion parameters when they are exposed to such hygrothermal environments. The diffusion coefficient is a crucial hygrothermal response parameter used for theoretical estimation of ease of moisture absorption in a material. The hygrothermal characterisation of polymer composites involves long term exposure of their specimens to climatic chambers maintained at intended temperature and humidity conditions.

Much work has been done in the area of the prediction of moisture content (or concentration) in a composite [1]. Theoretical models based either on one dimensional Fickian diffusion or 3D diffusion are used to determine the diffusion coefficient, D_c, as a function of the moisture content and the maximum moisture content (M_m) in the saturated state [1,2].

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0731-6844/99/10 0942–12 $10.00/0

A recent review by Wolff discussed the effects of moisture on advanced composite materials used primarily in the aerospace industry [3].

Diffusion coefficients (i.e., diffusivity of the material in the direction of the diffusion) for glass fiber epoxy composites containing different layups of chopped fiber and woven fabrics have been experimentally determined by Singh, Rao, et al. [4]. Diffusion coefficients for laminates with chopped strand mats at the surface were found to be considerably higher than those with fabrics [5].

Diffusion coefficients data for a variety of (aramid/epoxy, carbon/epoxy and glass/epoxy) composites have been reported by Aditya and Sinha [6].

EXPERIMENTAL DETAILS

Materials and Specimen Preparation

Test laminates were made out of woven glass fabric (CS Interglass, 8 MIL thick)/epoxy (LY5052 RT cure system of Ciba Geigy) composite systems in the present work. The specimens were cut from post cured (85°C/2 hrs) laminates fabricated out of stackings (warp on warp) of pre-impregnated layers room temperature cured under a standard vacuum bag molding (RTVBM) procedure, inhouse developed. The fiber weight fraction of all the laminates was maintained at 65 ± 3% (or 0.5 V_f). Edge coated test specimens for Longitudinal Tension (LT), Transverse Tension (TT), Longitudinal Compression (LC), Transverse Compression (TC) and In-plane Shear (IPS) tests were cut from the laminates, the thickness of each specimen being 2 mm (±0.1 mm). The tensile test specimen dimensions were 250 mm × 25 mm × 2 mm and the compressive test specimen dimensions were 120 mm × 12.6 mm × 2 mm. IPS test specimen dimensions were 130 mm × 25 mm × 2 mm.

A large number of specimens were chosen to comply with the acceptance requirements of composites for reliable estimation of their long term performance for aeronautical application. A total of 90 specimens classified as LT, TT, LC, TC and IPS were used for hygrothermal conditioning. Three sets of six specimens for each type of test were subjected to exposure for the three conditions.

ENVIRONMENTAL CONDITIONING

All test specimens in bone dried state (already oven post cured at 85°C/2 hrs) were weighed for their initial weights and grouped into three sets. One set was conditioned at 45°C/85% RH, the second set at 45°C/95% RH and the third set of specimens at 45°C/immersion (100% RH).

The specimens were weighed periodically (at least twice a week) starting from the instant the specimens were placed in the chamber. The weight gains of specimens were recorded as a function of time which in turn was expressed as the moisture gain of the specimens.

The percentage moisture content is recorded throughout this paper as percent of dry weight [7–9] according to the following expression

$$\% \text{ Moisture gain} = \frac{\text{Wt. of specimen at any time} - \text{Wt. of bone dry specimen}}{\text{Wt. of bone dry specimen}} \times 100$$

The % moisture gain values of each specimen were plotted against sq. root of time in hours to obtain the moisture gain curves. Initially, all the curves show linearity, the slope being proportional to the diffusivity of the material. After an extended time, the curves approach asymptotically the maximum or saturation moisture level M_m. Also the time dependent parameter G, defined as the ratio between the moisture weight gain at time t and the moisture weight gain at equilibrium (with zero initial moisture content) is plotted against the dimensionless diffusion parameter ($D_c t/h^2$) for all cases.

The diffusion coefficients were calculated from the initial slope of the curve according to the relationship,

$$D_c = \pi [h / 4M_m]^2 \left[M_2 - M_1 / \sqrt{t_2} - \sqrt{t_1} \right]^2 \quad (1)$$

where M_m is maximum moisture content, M_1 is the moisture content at time t_1, M_2 is the moisture content at time t_2, and h is the specimen thickness.

Fickian Diffusion Model and Correlations

The moisture absorption by a solid material [7,10] can be predicted based on the classical theory of diffusion described by Fick's second law, that at time t,

$$\delta c/\delta t = \delta/\delta x [D_c \delta C/\delta x] \quad (2)$$

where C is the moisture concentration, D_c is the diffusion coefficient, and x is the space coordinate in the direction normal to the section.

For composites using epoxy matrices, the diffusion coefficient is by and large concentration independent and hence of the through thickness location, so the above equation can be written as

$$\delta c/\delta t = D_c (\delta^2 c/\delta x^2) \quad (3)$$

where D_c is diffusion coefficient.

Considering a bone dry specimen ($M_i = 0$) and noting that moisture content (M_i) can be expressed as

$$M_i = A \int c \, dx \quad (4)$$

The solution of Equation (3) is given by Jost [11]

$$G = \frac{M_t}{M_m} = 1 - \left[\frac{8}{\pi}\right] \sum_{j=1}^{D_0} \frac{\exp[-(2j+1)^2 \pi^2 (D_c t / h^2)]}{(2j+1)^2} \quad (3)$$

where a plot of dimensionless absorption parameter $G = (M_t/M_m)$ against the dimensionless diffusion parameter $(D_c t/h^2)$ represents the Fickian diffusion correlation for the composite. A good match of the experimental data with this curve therefore confirms the Fickian model for the material considered.

RESULTS AND DISCUSSION

Determination of Diffusion Parameters (M_m, D_c)

Figures 1–3 show plots of % moisture absorption vs square root of time in hours for LT, TT, LC, TC and IPS test specimens of glass/epoxy conditioned at 45°C/85% RH, 45°C/95% RH and 45°C/immersion. The nature of the curves indicate the Fickian diffusion pattern with an initial linear portion and thereafter remaining concave to time axis until maximum moisture content (M_m) is reached. The D_c values for all specimens were calculated using Equation (1). The averaged D_c values, for a given type of test specimen were also obtained. The details of results for the test specimens conditioned are given in Table 1.

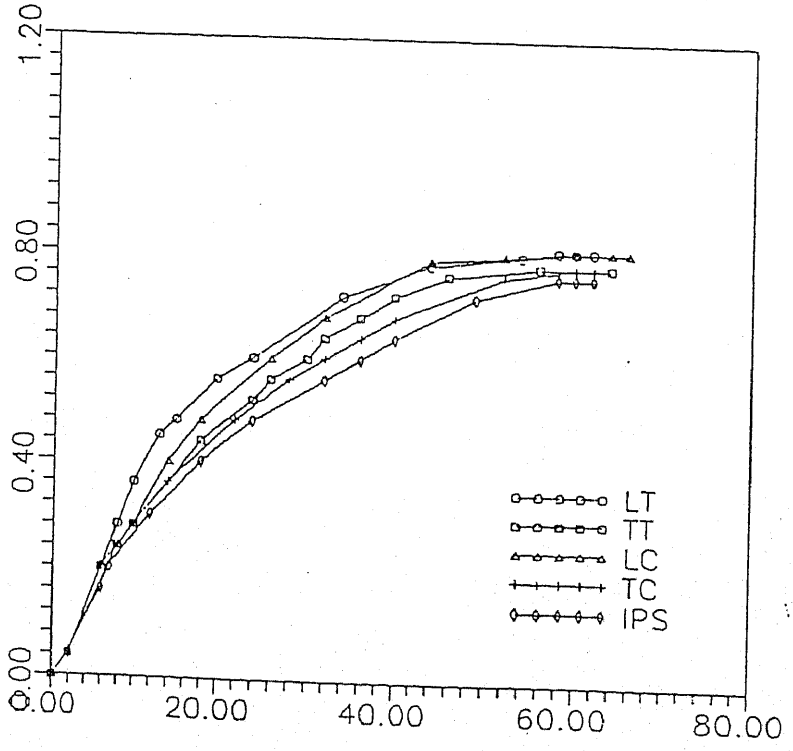

Figure 1. Moisture absorption curves for glass/epoxy test specimens conditioned at 45°C/85% RH: x-axis: sq. root of time in hrs and y-axis: % moisture gain.

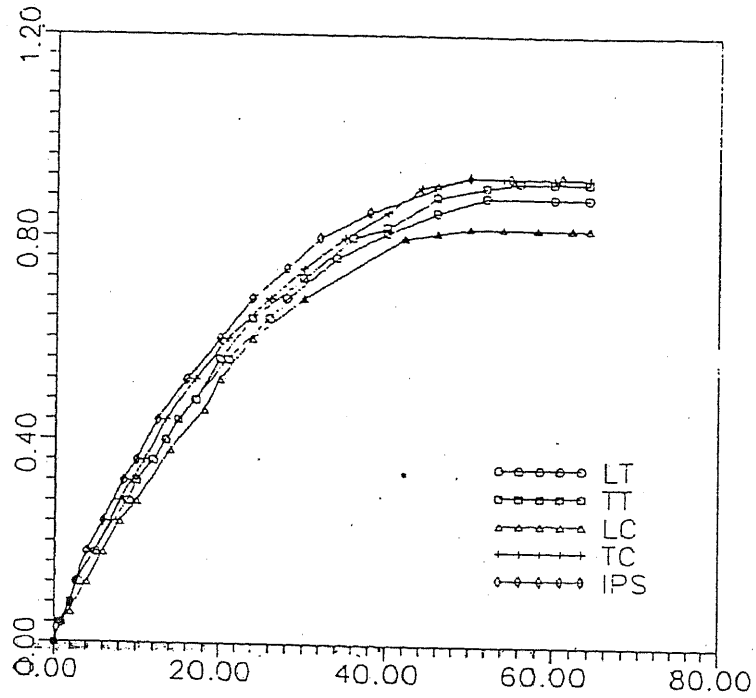

Figure 2. Moisture absorption curves for glass/epoxy test specimens conditioned at 45°C/95% RH: x-axis: sq. root of time in hrs and y-axis: % moisture gain.

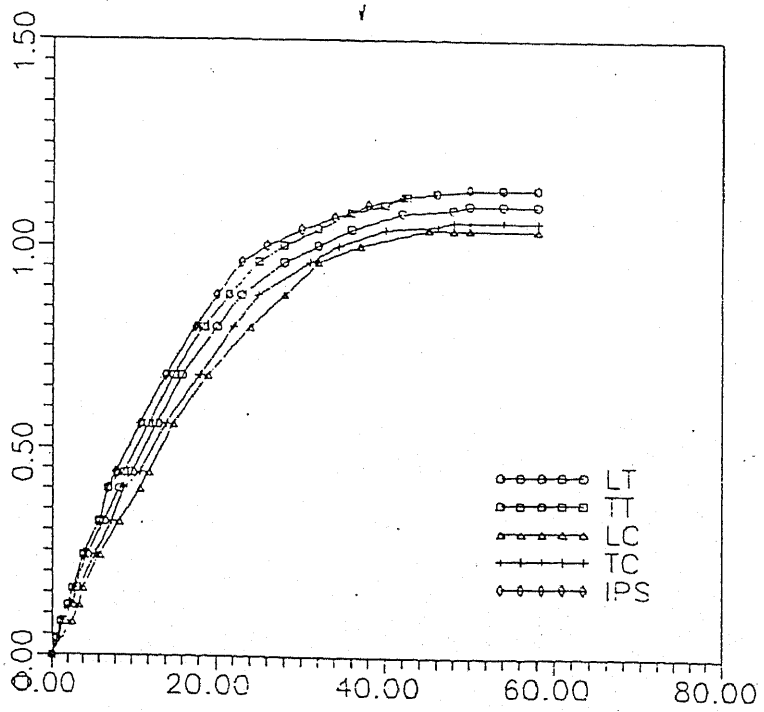

Figure 3. Moisture absorption curves for glass/epoxy test specimens conditioned at 45°C/immersion: x-axis: sq. root of time in hrs and y-axis: % moisture gain.

Table 1. Comparison of diffusion parameters for different glass/epoxy test specimens conditioned at 45°C/85% RH, 45°C/95% RH, 45°C/immersion.

Test Specimen	(%) M_m at			$D_c \times 10^{-7}$ mm²/sec		
	85% RH	95% RH	Immersion	85% RH	95% RH	Immersion
LT	0.80	0.88	1.10	5.45	4.50	2.83
TT	0.78	0.91	1.14	6.63	4.87	3.10
LC	0.75	0.84	1.04	7.75	6.18	4.05
TC	0.76	0.92	1.06	8.56	5.84	4.40
IPS	0.79	0.92	1.14	6.98	5.15	3.35

Also for the same thickness (i.e., same no. of layers), and for different test specimens of the same laminate under one set of test conditions, a small variation in the diffusivity values is noticeable. For a given material there are variations in the diffusivity values reported from one investigator to the other. The likely reason for these variations is the differences in the curing processes used in the different laboratories. It is very evident that even slight, unintentional differences in the curing process may alter the value of D_c significantly [12].

The M8 gain [13,14] of Springer was applied for the 45°C/95% RH conditioning case and found to yield M_m, T_m and D_c values comparable to experimental values obtained (Table 2).

Fickian Correlation Curves

Figures 4–6 show the correlation curves for all types of test specimens subjected to the three exposure conditions studied. Figures 4–6 indicate fairly good correlation between the analytical and experimental for all the cases of conditioning.

Table 2. Comparison of analytical and experimental values obtained for glass/epoxy specimens conditioned at 45°C/95% RH.

Test Specimen	$D_c \times 10^{-7}$ mm²/sec		T_m (in days)		
	Analytical*	Experimental	Analytical	Experimental	% M_m
LT	3.78	4.50	89	84	0.88
TT	4.75	4.87	99	85	0.91
LC	5.92	6.18	84	85	0.84
TC	4.85	5.84	97	78	0.92
IPS	5.33	5.15	90	85	0.92

*Values obtained using M8 gain of Springer [8,9].

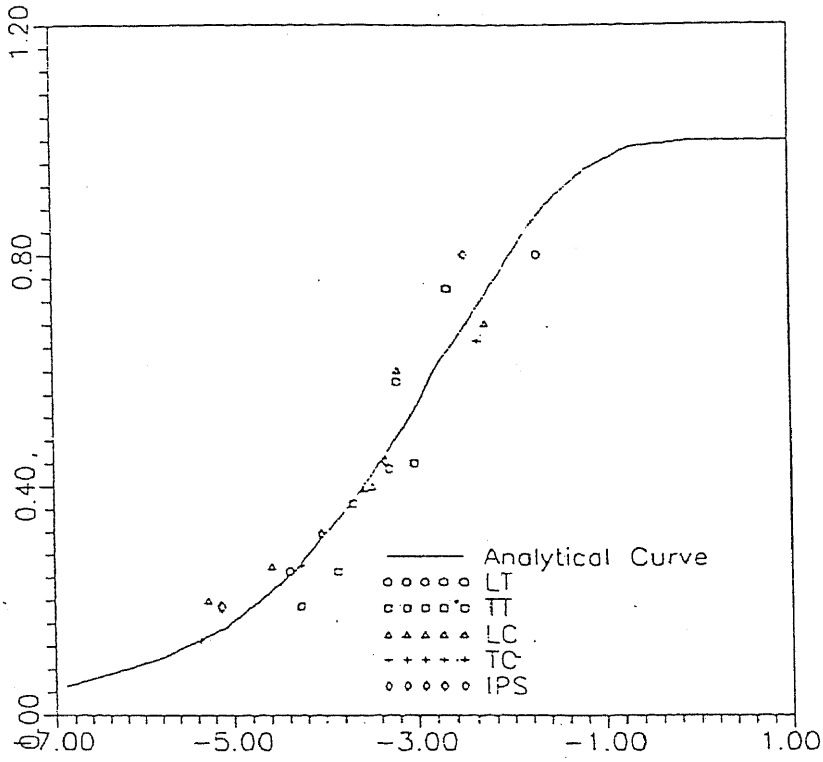

Figure 4. Comparison of analytical and experimental values for glass/epoxy test specimens conditioned at 45°C/85% RH: x-axis: ln $(D_c\, t/h^2)$ *and y-axis:* $G = (M_t/M_m)$.

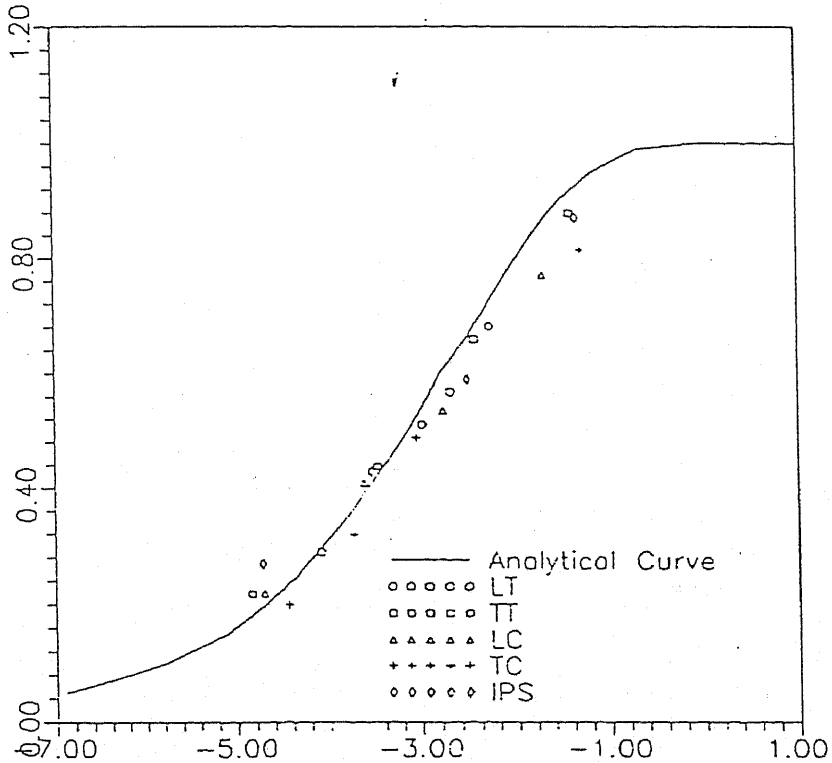

Figure 5. Comparison of analytical and experimental values for glass/epoxy test specimens conditioned at 45°C/95% RH: x-axis: ln $(D_c\, t/h^2)$ *and y-axis:* $G = (M_t/M_m)$.

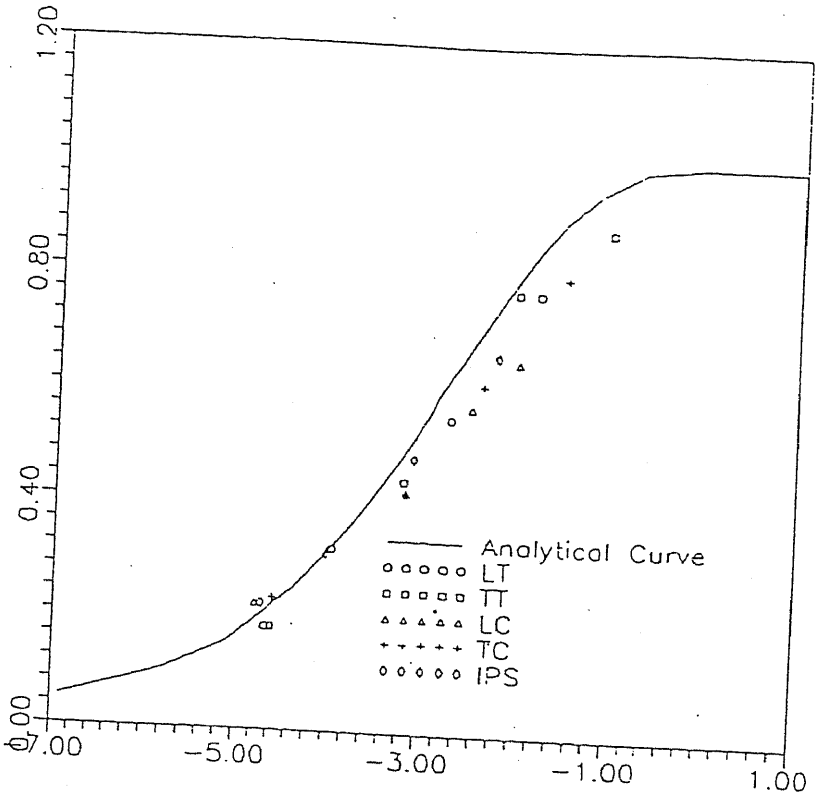

Figure 6. Comparison of analytical and experimental values for glass/epoxy test specimens conditioned at 45°C/immersion: x-axis: ln (D_c t/h^2) and y-axis: G = (M_t/M_m).

Variation of Maximum Moisture Content (M_m) with % RH

It is well known that the maximum moisture content is a strong function of relative humidity (RH) of the ambient. For a material immersed in liquid, M_m is a constant

$$M_m = \text{constant (liquid)}$$

For a material exposed to humid air, M_m depends on the relative humidity (ϕ), according to the relationship

$$M_m = a(\phi)^b \text{ (humid air)}$$

$$\log M_m = \log a + b \log (\phi) \tag{6}$$

From the data obtained at two different conditions, viz., 45°C/85% RH and 45°C/95% RH, a and b were evaluated for the above equations as $a = 0.954$ and $b = 1.21$.

Figure 7 shows the variation of M_m with % RH at constant temperature in general for a glass/epoxy system.

Also, Figures 8–12 show the variation of $\%M_m$ with percentage relative humidity for LT, TT, LC, TC and IPS specimens individually.

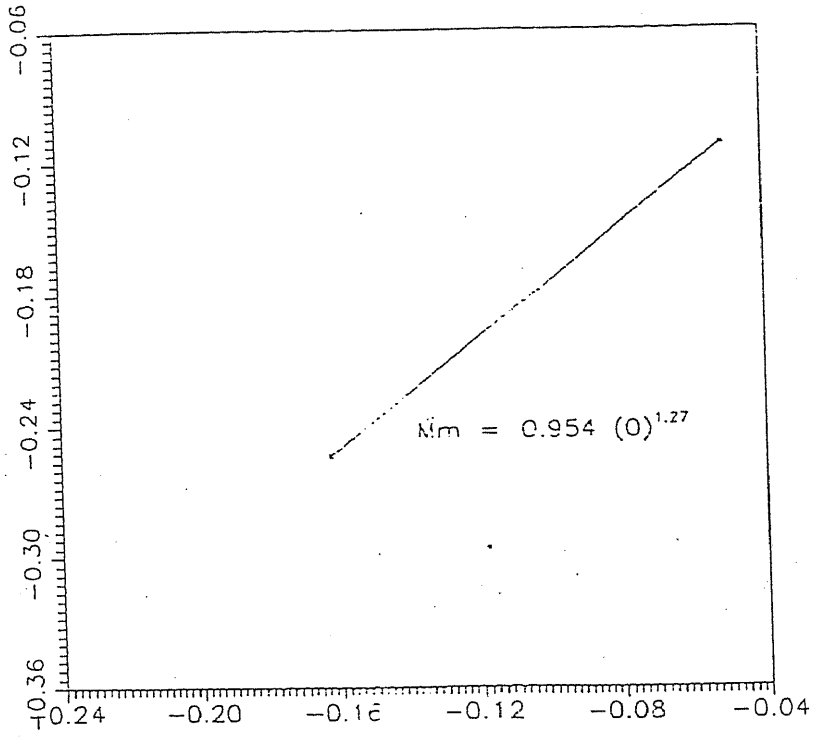

Figure 7. Variation of maximum moisture content with % relative humidity for a glass/epoxy system.

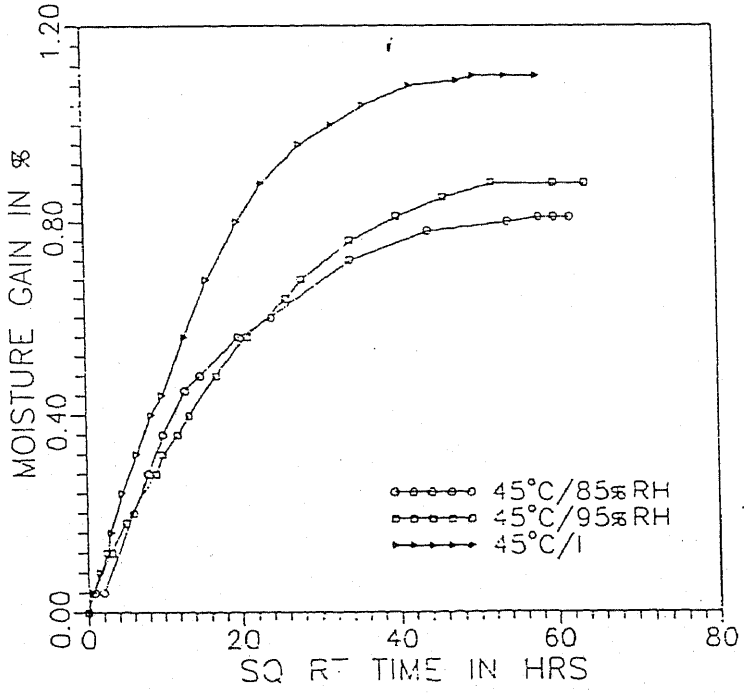

Figure 8. Variation of maximum moisture content with relative humidity for LT specimens of glass/epoxy system: x-axis: sq. root of time in hrs and y-axis: % moisture gain.

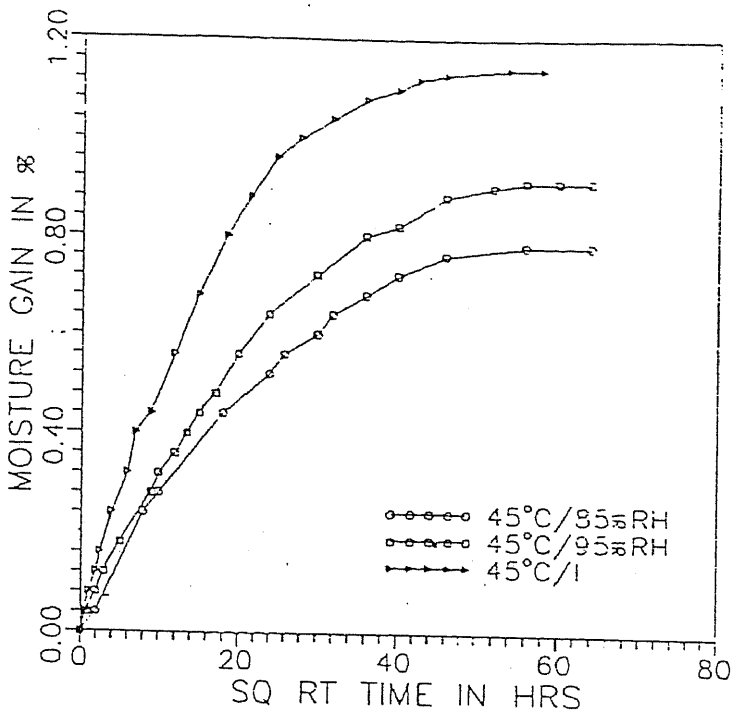

Figure 9. Variation of maximum moisture content with relative humidity for TT specimens of a glass/epoxy system: x-axis: sq. root of time in hrs and y-axis: % moisture gain.

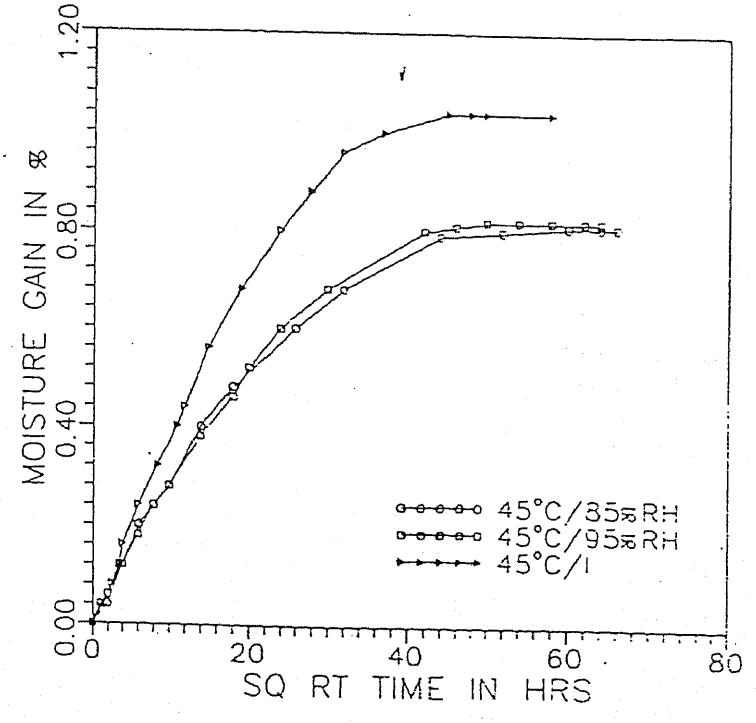

Figure 10. Variation of maximum moisture content with relative humidity for LC specimens of a glass/epoxy system: x-axis: sq. root of time in hrs and y-axis: % moisture gain.

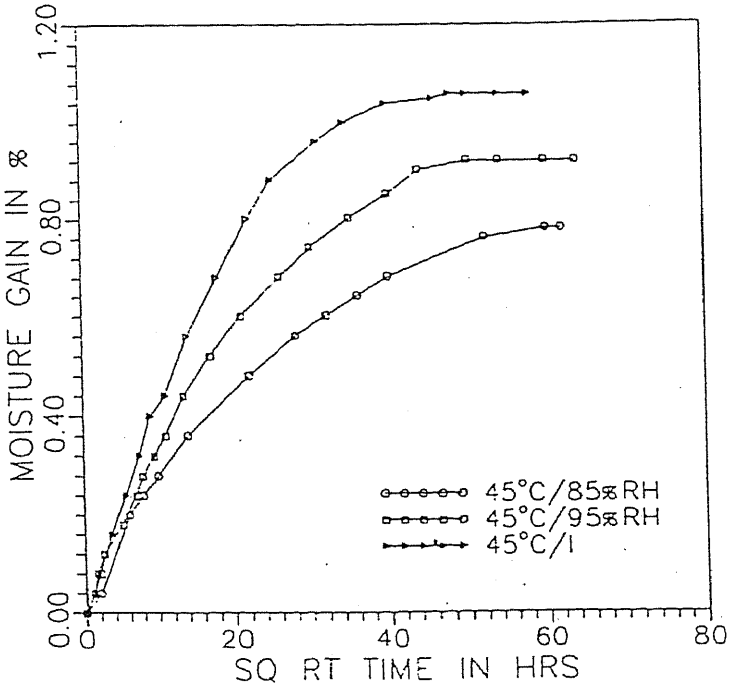

Figure 11. Variation of maximum moisture content with relative humidity for TC specimens of a glass/epoxy system.

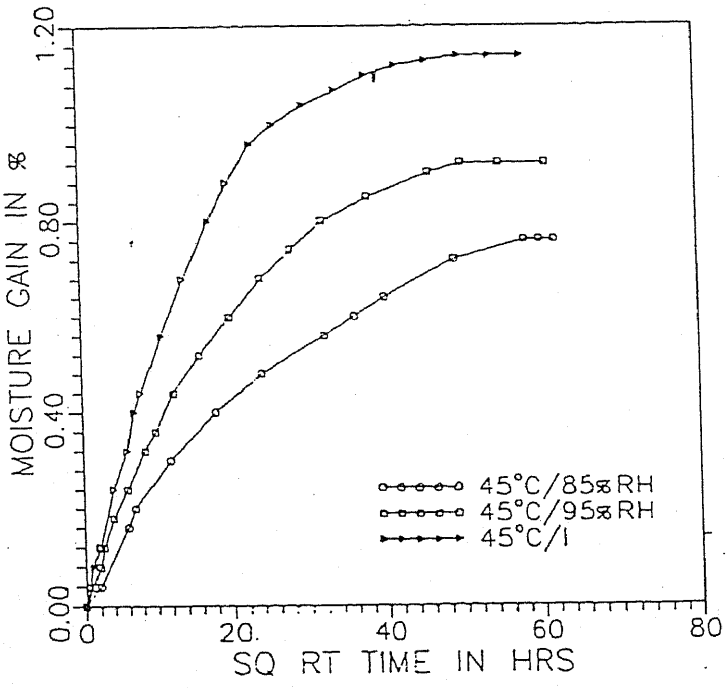

Figure 12. Variation of maximum moisture content with relative humidity for IPS specimens of a glass/epoxy system.

CONCLUSIONS

- The bidirectional glass/epoxy specimens exhibited the Fickian behavior at different hygrothermal conditions studied.
- The maximum moisture content increased with the increase in relative humidity and the diffusion coefficient (D_c) values decreased.
- The diffusion coefficient (D_c) values for a given specimen marginally decrease with an increase in relative humidity.
- The experimental values for D_c and T_m compared well with those predicted analytically.
- For the same resin system under similar test conditions the D_c values for glass/epoxy is in general higher than those obtained for carbon/epoxy specimens.

ACKNOWLEDGMENTS

The authors are thankful to Dr. T. S. Prahalad, Director, National Aerospace Laboratories for all the support received during the investigations. They also acknowledge the assistance rendered from Mr. Rajendra Prakash and Mr. Azar Amir in the preparation of test specimens.

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